Effective intersublattice exchange couplings in rare-earth–transition-metal intermetallics from an *ab initio* calculation

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A method is outlined for the *ab initio* determination of effective intersublattice exchange couplings J_{RT} in rare-earth–transition-metal intermetallics. The results obtained for $R_2Fe_{14}B$ ($R =$ Gd,Tb,Dy) and Ho_2Fe_{17} by the linear-muffin-tin-orbital theory in atomic sphere approximation are in excellent agreement with experimental data. It is shown that J_{RT} is not exclusively determined by the exchange-correlation part, but also strongly by other contributions to the total energy.

Rare-earth–transition-metal intermetallics are of high technological importance because of their excellent hard magnetic properties.[1-3] For the development of new materials a better understanding of the physical mechanisms, which determine the intrinsic magnetic properties (high saturation magnetization, magnetic anisotropy, and Curie temperature) are of invaluable importance. In recent publications, *ab initio* results have been reported for the local magnetic moments and hyperfine fields[4] as well as for the magnetic anisotropy[5,6] of $R_2Fe_{14}B$ with $R =$ Nd, Gd, Tb, Dy, Ho, and Er. This paper is concerned with a calculation of exchange interactions, which are related to the Curie temperature.

Most experimentalists discuss the magnetic properties of rare-earth intermetallics within the framework of a two-sublattice model[1,2,7] of a rare-earth (R) and a transition-metal (T) sublattice[8] coupled via effective exchange interactions J_{RT}. Two different microscopic models for the exchange coupling have been proposed in the literature. In both models it is emphasized that because of the localized nature of the $4f$ states the interaction between the $4f$ spin and an itinerant electron spin can be mediated only through the local exchange interaction on the rare-earth atom, mainly by the intra-atomic exchange $J_{4f\text{-}5d}$, the R $5d$ moment being induced by the R $4f$ spin density. In the model of Campbell,[9] the inter-sublattice coupling is generated by a subsequent direct $5d\text{-}nd$ interaction with any other nd spin moment. In contrast, Brooks and co-workers[10,11] have outlined that for RFe_2 the $5d$ polarization by the $4f$ spin density occurs only if there is a $3d\text{-}5d$ hybridization. They stated that the intersublattice coupling is not via a $4f\text{-}5d\text{-}3d$ exchange path, but arises from a combination of local $4f\text{-}5d$ exchange and $5d\text{-}3d$ hybridization. Calculations on both of these two lines[10-13] (each requiring several approximations) surprisingly yield results in fair agreement with experiments, although the underlying physical mechanisms are quite different.

In this Rapid Communication we report on calculations that do not rely on specific models for exchange interactions in rare-earth intermetallics, but that determine from first principles the effective intersublattice interaction J_{RT} in the spirit of the phenomenological two-sublattice model. In this model the two sublattices are represented by the $4f$ moments of the R atoms (any $5d$ contribution is not considered explicitly) and the magnetic moments of the Fe atoms. The change of the total energy upon disalignment of the two sublattices is ascribed to an effective exchange interaction J_{RT} between the corresponding spin moments. Any intra-atomic contributions arising from a possible change of the $5d$ moments (see below) are not considered explicitly but are integrated in J_{RT}. To proceed along this line, we have calculated the change in total energy achieved by rotating the $4f$ moments out of the ground-state orientation by an angle of 180°. This method for a calculation of exchange interactions has been already used for transition metals; see for instance Ref. 14. Thereby, problems may arise because the magnitude of transition-metal moments may

TABLE I. The local magnetic spin moments for the two crystallographically inequivalent Gd sites, the six Fe sites, and the B site in $Gd_2Fe_{14}B$ for the ground-state (antiferromagnetic) orientation of the Gd and Fe sublattices and for the ferromagnetic orientation.

Local magnetic spin moments		Ground state	Reversed $4f$ spin
Gd 4(g) total		−7.68	6.76
	$4f$	−7.00	7.00
	d	−0.49	−0.15
	p	−0.13	−0.07
	s	−0.05	−0.02
Gd 4(f) total		−7.64	6.79
	$4f$	−7.00	7.00
	d	−0.45	−0.13
	p	−0.13	−0.07
	s	−0.06	−0.02
Fe 4 (e)		2.04	2.02
Fe 4(c)		2.48	2.40
Fe 8(j_1)		2.33	2.24
Fe 8(j_2)		2.60	2.58
Fe 16(k_1)		2.19	2.15
Fe 16(k_2)		2.39	2.34
B 4(f)		−0.18	−0.19
$\bar{S}_{Fe}$		1.17	1.14

TABLE II. The changes upon $4f$ spin reversion of the total energy ΔE, and of the exchange-correlation energy ΔE_{xc}, of the unit cell.

	ΔE [mRy]	ΔE_{xc} [mRy]
$Gd_2Fe_{14}B$	152	535
$Tb_2Fe_{14}B$	116	423
$Dy_2Fe_{14}B$	85.3	319
Ho_2Fe_{17}	33.6	179

change drastically when going from a ferromagnetic to an antiferromagnetic orientation,[15] which necessitates the use of rather small rotation angles.[16−18] In contrast, in our case the Fe moments remain remarkably constant (see below), and the $4f$ moments of the R atoms (which most probably will not change noticeably in reality because they arise from deep core states) are kept constant in our calculational method (see below).

Our calculated change of the total energy ΔE per unit cell is represented in the two sublattice model by a molecular field B_{RT}^m for the spin S_R of the R atom, or by a related effective intersublattice exchange parameter J_{RT}:[19]

$$\Delta E = -4\mu_B B_{RT}^m S_R = -4J_{RT} S_R \bar{S}_T Z_{RT} Z_R . \qquad (1)$$

Here μ_B denotes Bohr's magneton, Z_R is the number of R atoms in the unit cell, Z_{RT} denotes the number of nearest-neighbor T atoms of an R atom and $\bar{S}_T$ is the effective transition-metal spin moment, averaged over all T sites in the crystal.

To summarize, the central point of this two-sublattice model is that the change upon $4f$ spin reversion of *any* contribution to the total energy is attributed to the effective intersublattice interaction parameter J_{RT}, not only the change of the exchange contribution as in the models of Refs. 10–13.

The scalar-relativistic calculations are based on the local-spin-density approximation and the linear-muffin-tin-orbital theory in atomic sphere approximation,[20] including the combined correction term. The $4f$ states are treated as open core states with a Hund's rule restriction for the $4f$ spin, according to Brooks, Nordström, and Johansson.[11]

We now discuss our results for $R_2Fe_{14}B$ with $R =$ Gd,Tb,Dy and for Ho_2Fe_{17}. It becomes obvious from Table I that the magnetic moments on the various Fe sites indeed change only very slightly when reversing the $4f$ moment. We present the data for the case of $Gd_2Fe_{14}B$, for which the changes are strongest. There is a noticeable contribution of p and s states to the valence moment at the R site, which in turn is small compared to the $4f$ moment. Note that the valence contributions to

TABLE III. Comparison of our theoretical and the experimental data for J_{RT}.

	J_{RT} $[-10^{-22}$ J]	
$Gd_2Fe_{14}B$	1.46	This work
	1.46	Ref. 21
$Tb_2Fe_{14}B$	1.30	This work
	1.28−1.29	Ref. 21
$Dy_2Fe_{14}B$	1.15	This work
	0.88−1.16	Ref. 21
Ho_2Fe_{17}	1.17	This work
	1.2−1.4	Ref. 22

the total moments of the R sites change by a factor of about 2 upon reversion of the $4f$ spin moments. We do not have to worry about the related intra-atomic exchange contribution because this is attributed to the effective intersublattice coupling in the spirit of the two-sublattice model discussed above.

Table II represents the changes of the total energies, ΔE, and of the exchange-correlation part, ΔE_{xc}, upon $4f$ spin reversion. (It should be noted that the data for ΔE_{xc} are possibly not yet totally converged with respect to the number of irreducible k points,[14] although the data for ΔE are, and they are less accurate concerning convergence to self-consistency because only the total energy is variational.) In agreement with the findings of Peng and Jansen,[14] ΔE_{xc} is rather large but shielded by the system so that the change ΔE of the total energy is smaller than ΔE_{xc}. The value of J_{RT} thus would be overestimated if it were attributed to ΔE_{xc}. In contrast, relating J_{RT} to the change of the total energy according to (1) yields excellent agreement (Table III) with the most recent experimental data by Loewenhaupt *et al.*[21] for $R_2Fe_{14}B$ and Gubbens, van der Kraan, and Buschow[22] for Ho_2Fe_{17}.

To conclude, it has been shown that the effective intersublattice exchange interaction J_{RT} for rare-earth intermetallics can be determined by *ab initio* calculations within the framework of the local-spin-density approximation without relying on any model assumption, if J_{RT} is attributed to the change of the total energy upon $4f$ spin reversion, in analogy to the spirit of the two-sublattice model commonly used for an interpretation of experimental data.

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